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Abstract

The spin-resolved electronic band structure of Au-induced metallic atomic wires on a vicinal silicon surface, Si(553), was investigated using spin- and angle-resolved photoelectron spectroscopy. We directly measured the spin polarization of three partially filled one-dimensional metallic bands, a one-third-filled band, and the doublet of nearly half-filled bands. For the half-filled doublet, the strong apparent spin polarization was observed near the Fermi energy with a minor out-of-plane spin component. This observation is consistent with the Rashba-type spin-orbit splitting and with a recent experiment on a similar doublet of Si(557)-Au. In contrast, the one-third-filled band does not show a substantial spin polarization within the experimental accuracy, indicating a much smaller spin splitting, if any. These results are discussed for the origin of the partially filled bands and for the intriguing broken-symmetry ground state observed at low temperature.

Keywords: atomic wires, Rashba effect, spin- and angle-resolved photoelectron spectroscopy, 1D metal

1. Introduction

Spintronics is seeking high performance electronic devices that exploit the spin degree of freedom of electrons [1]. The spin–orbit coupling (SOC) is key in the physics of the methods used for manipulating electron spins by electric field. A spin field-effect transistor using the spin precession based on SOC was introduced [2]. A few materials systems with strong SOC, for recent examples, such as topological insulators [3] and heavy metal thin films [4, 5], were proposed as candidates for such spintronic applications. In these materials, the spin polarization emerges without external magnetic fields, even at room temperature, through the quantum spin Hall effect or the Rashba spin splitting of surface-state bands.

In particular, the Rashba effect [6], which is the lifted spin degeneracy due to the breakdown of the space inversion symmetry at crystal surfaces, has been actively studied in a few surface systems [4, 5]. However, most of the previous works studied metallic surface states on metal substrates or non-metallic surface states on semiconductors, for which a well-isolated spin transport channel is hardly realized. A metallic surface state with the spin splitting on a metal/semiconductor system, which was observed very recently, is thus important to overcome this limitation [7]. However, this metallic Rashba channel has only a moderate spin splitting. On the other hand, one-dimensional (1D) systems with small SOC, such as carbon nanotubes (CNTs) [8] or semiconductor quantum wires [9], have attracted attention because of their obvious merits in manipulating spin currents. Thus, 1D metallic systems with a large Rashba spin splitting on semiconducting substrates, if realized, would provide an exciting opportunity in this line of research. We recently found a 1D system with a large Rashba splitting on a Si surface, Pt-induced nanowires on Si(110) [10]. However, this system is not metallic and a 1D metallic system with a substantial Rashba splitting remains to be discovered.

The metallic atomic wires on vicinal Si surfaces induced by Au adsorbates, such as Si (553)-Au, Si(557)-Au, and Si(5 5 12)-Au, are candidates for such spin-split 1D metallic systems. Scanning tunneling microscopy (STM) and angle-resolved photoelectron spectroscopy (ARPES) studies showed the formation of regular atomic wire arrays along step edges and 1D electronic structures with multiple metallic bands, respectively (see figure 1 for the atomic structure model schematics and the band structure) [11–13]. These systems share two closely lying nearly half-filled bands [12, 14–16], whose origin has been under debate. The doublet band was first attributed to the spin-charge separation of a Tomonaga–Luttinger liquid [11]. However, the following higher-resolution ARPES studies clearly ruled out this possibility [15, 17] and further suggested that the two bands originate from different atomic rows on the surfaces [15, 16]. Recently, this characteristic doublet in Si(557)-Au was theoretically suggested as spin-split bands by the Rashba effect on Au atomic chains [18] and a similar calculation for the Si(553)-Au surface followed [19]. The subsequent ARPES experiment interpreted the fine intensity modulation of the doublet in Si(553)-Au based on the Rashba effect [20]. Very recently, the spin splitting was confirmed directly for the half-filled doublet of Si(557)-Au using spin- and angle-resolved photoelectron spectroscopy (SARPES) measurement [21]. However, this system has a significantly large density of defects, mainly because of the Si adatoms on terrace sites [13, 28]. In this respect, the Si(553)-Au surface is important because it has no Si adatoms and thus can be prepared with an extremely low defect density within a very good vacuum condition [13, 28]. Nevertheless, no direct experimental measurement of the spin polarization for Si(553)-Au was reported so far.

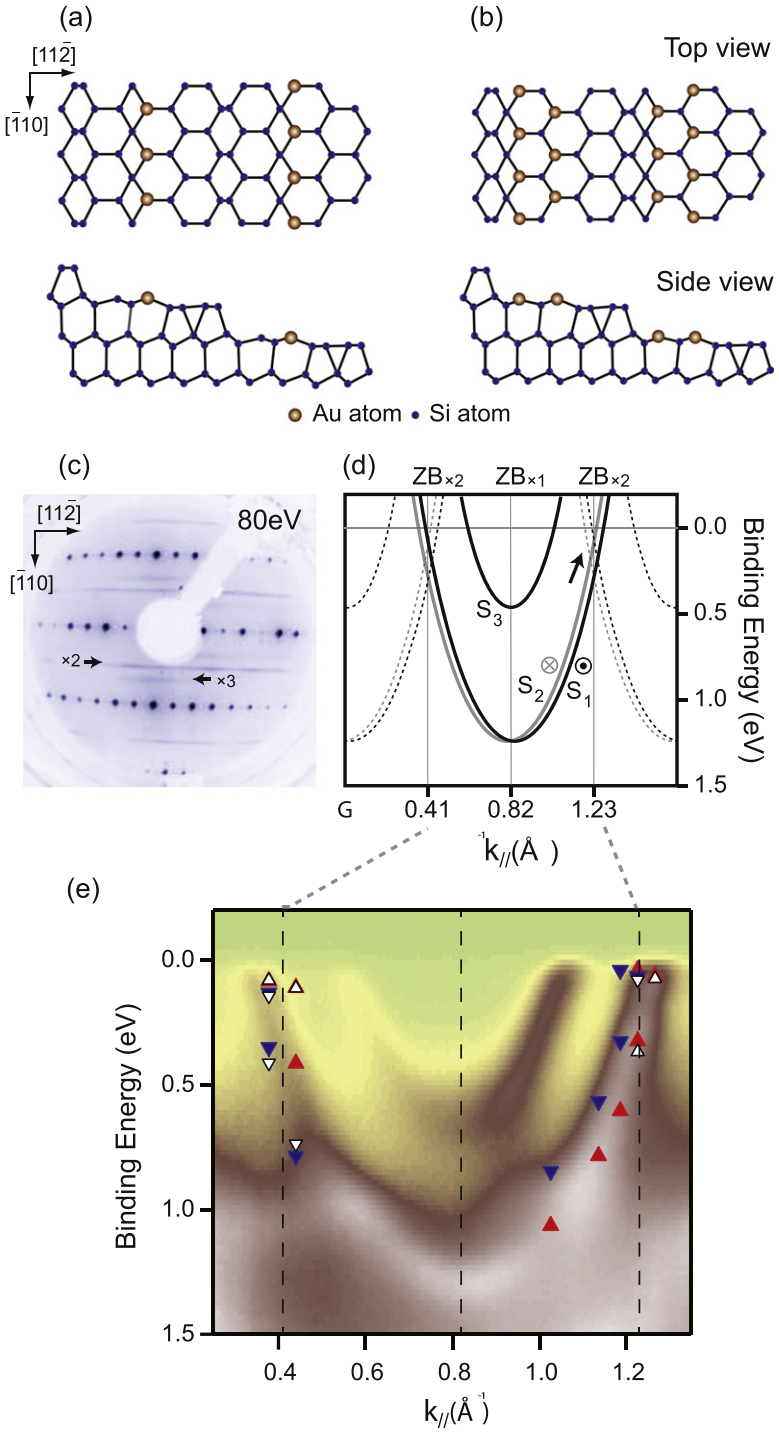

Figure 1. Schematic illustrations of two structural models for Au atomic wires on the Si (553) surface; (a) the single-row and (b) double-row model. (c) LEED pattern of the Si (553)-Au surface with a well-ordered wire array taken at 120 K with an electron energy of 80 eV. (d) Schematics of the measured band structure of this surface. $ZB_{\times 1}$ and $ZB_{\times 2}$ indicate the Brillouin zone boundary for the ×1 and ×2 unit cells along the wire. (e) Corresponding spin-integrated band dispersions measured. The triangles in (e) are the peak positions determined in the SARPES measurements discussed next.

Moreover, the spin structure of Si(553)-Au is even more intriguing because of an antiferromagnetic ground state predicted theoretically along the step-edge Si chains [22] and because of the existence of an additional metallic one-third-filled band. It is also noteworthy that two different lattice distortions, that is, the double (×2) and triple (×3) period ones, were observed on this surface at low temperature and are related to nearly half- and one-third-filled bands, respectively, through the Peierls instability mechanism [16, 23–25]. In contrast to this mechanism, very recently, the absence of the band gap for the low temperature phase was indicated [26, 27]. Because the spin polarization of the metallic band would definitely affect the Peierls instability, the direct measurement of spin becomes even more important for this intriguing system.

In this work, we directly measured the spin polarization using SARPES of the three metallic surface-state bands on the Si(553)-Au surface, which exhibits a relatively well-ordered wire array with less defects than Si(557)-Au [13, 28]. The Rashba spin splitting of the half-filled doublet is unambiguously supported. In contrast, the spin splitting of the third band is, if present, shown to be substantially smaller than that of the Rashba doublet. The implications of the present results for atomic origins of the 1D metallic bands, the structure models proposed recently, and the broken-symmetry ground states at low temperature are discussed.

2. Experimental

A clean Si(553) surface, 12.5° miscut from the [111] direction toward [11 $\bar{2}$], was prepared by flash heating up to 1520 K with sequential postannelings at lower temperatures [12, 13]. A Si (553)-Au surface with a well-ordered atomic wire array was fabricated in a way similar to previous reports; depositing Au on Si(553) held at 970 K with a postannealing at the same temperature [12, 13]. The clean Si(553) and well-ordered Si(553)-Au surfaces were characterized by low-energy-electron diffraction (LEED).

The SARPES measurements were performed at Hiroshima Synchrotron Radiation Center using undulator radiation (34 eV) using a high-resolution SARPES machine (ESPRESSO), which combines a high-performance hemispherical electron analyzer with two efficient spin detectors based on very-low-energy-electron diffraction from a magnetized target [29]. The instrumental angular and energy resolutions were set at 0.75° and 20 meV, respectively. In this system, we can obtain SARPES spectra for complete three-dimensional spin orientations; in-plane (x, y) and out-of-plane (z) components (see figure 2(a) and the insets of figures 2 and 3). The out-of-plane spin direction lies along z in the x_s and y_s plane with an angle defined by the sample-to-analyzer angle. The spin asymmetry A is defined as $A=[(I_{plus}-I_{minus})/(I_{plus}+I_{minus})]$, where I_{plus} and I_{minus} are the measured intensities of reflected electrons by the positively and negatively magnetized target, respectively. The spin polarization P is obtained as $P = A/S_{eff}$, where S_{eff} is the effective Sherman function. S_{eff} was calibrated as 0.265 using the spin polarization of a Bi(111) film [29]. High-resolution spin-integrated ARPES data were taken at the BL6U beamline of UVSOR synchrotron radiation facility (Okazakl, Japan) using photon energy of 46 eV at a temperature of 20 K.

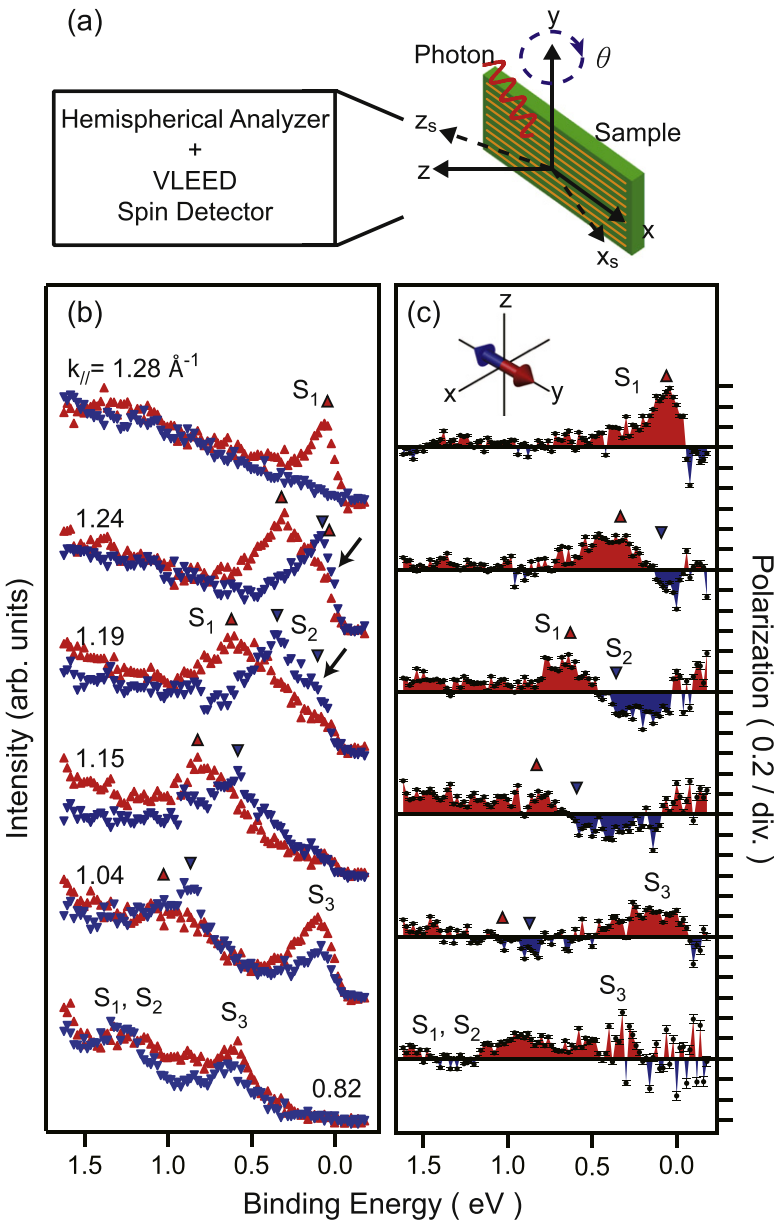

Figure 2. (a) Schematics of the SARPES measurement geometry. The electron analyzer and sample coordinates are indicated by (x, y, z) and (x_s, y_s, z_s), respectively. Note, x and x_s axes coincide. (b) The SARPES spectra and (c) the corresponding spin polarization along the wires (k_x) for the in-plane spin components. The spin direction of each spectrum is indicated in the inset. Note that, for the spectra in (c), the spin polarization lies along k_y.

3. Results and discussion

Figures 1(a) and (b) show the two most recently proposed atomic structural models of the Si (553)-Au surface [30]. They contain single and double rows of Au atoms, respectively, substituting surface Si atoms on terraces. The single-row model, adapted in earlier works, could reproduce the room-temperature STM images through the simulation using first-principles

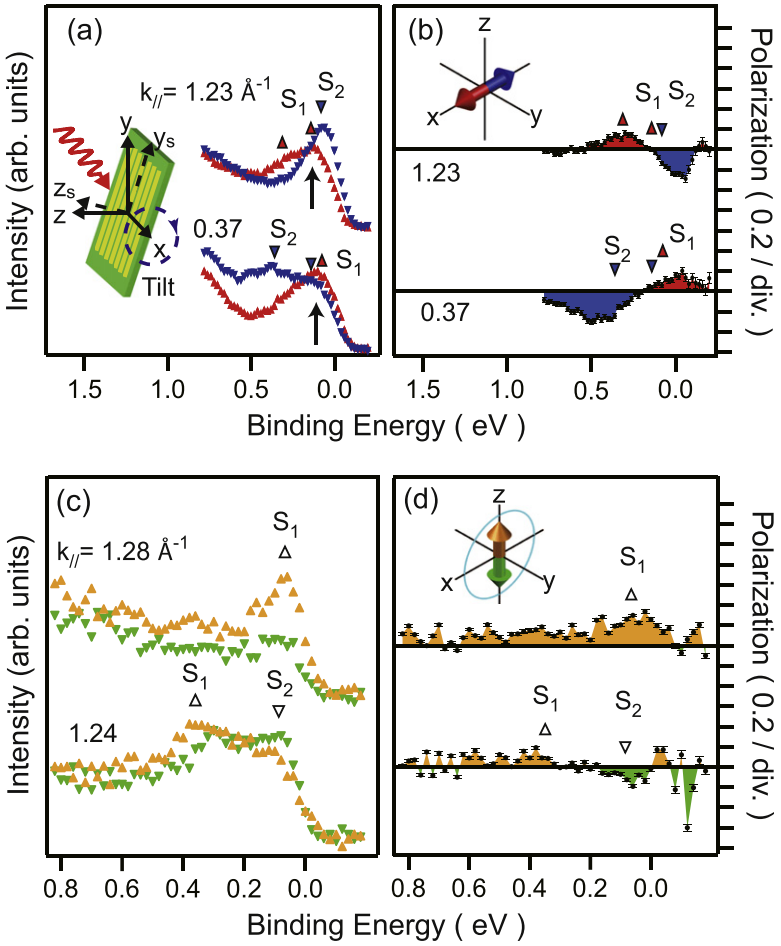

Figure 3. (a) and (b) Similar SARPES data as figure 2 but for in-plane spin polarization with spins along k_x for two particular k-points showing the S1/S2 doublet. (c) and (d) Similar SARPES data for the out-of-plane spin polarization.

density-functional theory calculations [13, 19]. However, it could not fully reproduce the electronic band structure measured experimentally [13, 19]. Figure 1(c) contains the schematics of the band dispersions reported previously; the doublet of half-filled bands is called S1 and S2 and the one-third-filled band is called S3 [12, 16]. The splitting between S1 and S2 is about 0.25 eV in energy and 0.04 $Å^{-1}$ in momentum near the Fermi energy. In the calculations based on the single Au row model, the S1/S2 and S3 bands are attributed to Au-Si chains on terraces and Si chains on step edges, respectively. As mentioned previously it appears that this provides a natural explanation for the two different distortions with the periodicity of ×2 and ×3 observed at low temperature (figure 1(c)) on Au-Si and Si step-edge chains, respectively, through the Peierls mechanism.

In contrast, a recent experimental study suggested two times higher Au coverage [31] and a subsequent density-functional theory calculation introduced the double-row model in figure 1(b) [32]. This model could better reproduce the band structure and the S1/S2 and S3 bands are all related to two neighboring Au-Si chains. The Rashba splitting origin of the S1 and S2 bands was also supported within this model [32]. However, the difference between the band structures based on some derivatives of the single-row model and the double-row model is not

too large [19, 30, 32]. Moreover, the agreement between the calculated and measured surface band dispersions is not conclusive enough to unambiguously determine the surface structure. In particular, an extra surface-state band due to Si step-edge chains is expected in the theory, but was not observed in the experiments [12, 16]. This model explains the low-temperature ×3 feature on Si step-edge chains as due to the antiferromagnetic spin ordering and the ×2 feature along Au-Si chains as due to the small structural distortion accompanying the spin polarization [22]. In fact, this is the first theoretical model explaining the low temperature superstructures observed. In this model, the one-third-filled band is not related to the ×3 phase transition at all and, thus, not expected to open a band gap. The metallic character of Au-Si chains was recently shown to persist into the low temperature phase by scanning tunneling spectroscopy (STS) [26, 27]. That is, the current model for the low temperature phase converges into a picture of metallic Au-Si chains with a ×2 structural distortion and insulating Si step-edge chains with a ×3 antiferromagnetic order.

We measured SARPES spectra along k_y near the Fermi level crossings of S1–S3 as shown in figures 2 and 3. The *k*-points sampled are shown in figure 1(e). The spectra shown in figures 2(b) and 3(a) (figures 2(d) and 3(b)) are the corresponding SARPES spectra (spin polarizations) of in-plane spin components. In SARPES, the splitting of S1 and S2 states is very clear, which clearly evidences the large spin polarization of these two states; the spectral peak positions (or the leading edge positions very close to the Fermi energy) are clearly different for the left and right spin components by about 0.25 eV in the spin-polarized spectra and are in good agreement with the previous spin-integrated measurements [12, 16]. Note that we measured the spin perpendicular to chains on the surface and the spin orientation is opposite between the two Fermi crossings at 0.4 and 1.2 Å^{-1} [figures 3(a) and (b)]. Moreover, the spin splitting vanishes at the bottom of the band (0.8 Å^{-1}, see figure 2). Near the Fermi level crossings of S1 and S2, extra weak spectral features appear due to the back folding of the bands caused by the ×2 superstructure (see the arrow and the dashed lines in figure 1(e)). This gives both spin components at $\text{ZB}_{\times 2}$ as indicated by arrows in figures 2 and 3. Thus, the spin texture is fully consistent with the Rashba spin-orbit splitting.

The maximum apparent in-plane spin polarization is as large as 0.7 near the Fermi energy and the remaining spin polarization of 0.3 or so is the out-of-plan component, as shown in figures 3(c) and (d). The out-of-plane spin component was also observed in the previous measurement for the half-filled doublet band of Si(557)-Au. In that case, the out-of-plane component even surpasses the in-plane polarization, deviating significantly from the simple Rashba picture, in which the spin is perpendicular to the electron momentum and resides within the surface plane. This unusual spin orientation was, however, reproduced using the theoretical calculation and understood based on the strong in-plane potential gradient felt by the electrons of Au adatoms [21]. The local atomic structure around Au adsorbates on Si(553) and Si(557) are different due to the presence of the double Au chains on Si(553) and the Si adatom chains on Si(557). This structural difference may explain the different weight of the out-of-plane spin polarization.

The present direct measurement of the spin polarization conclusively confirms the SOC origin of the S1/S2 doublet suggested by recent theoretical calculations [19, 22]. This rules out the possibility that these two metallic states originate from Si atoms at step edges [15, 16]. Such noticeable SOC is possible basically because this doublet originates from the Au-Si chains, with a large contribution from the Au 6*p* orbital. Note that there is a consensus on the atomic origin of this doublet over different structure models at present [19, 22]. The energy splitting was

calculated to be about 0.2 eV near the Fermi level [18], which is consistent with the previous spin-integrated ARPES [12, 16] and the present SAPRES measurements. It is worthwhile to note that these values are significantly larger than that of a surface state of the Au(111) surface, which is about 0.1 eV [34, 35]. The size of the Rashba splitting is determined by the integral of the potential gradient along the surface normal as multiplied by the charge density. Furthermore, the in-plane anisotropy of the surface potential could enhance this splitting [33].

It is notable that this Rashba splitting is far larger than other 1D conduction systems with the SOC such as CNT and InGaAs/InAlAs quantum wires, for which the energy scale of the Rashba splitting is as small as 0.1∼10 meV [8, 36]. The present 1D metallic system with such a huge Rashba splitting is thus very much encouraging for the effort toward novel spintronic applications. Further experiments, such as the nanoscale spin transport measurements on the present system, if possible, would provide important insight into the realization of spintronic devices based on SOC. It should also be noted that the present system is based on a silicon substrate, which makes it much more attractive for any practical device applications. In recent years, 1D wires with a substantial Rashba splitting attract further attention because they, in the proximity with superconductors, can be used to realize a Majorana Fermion [37]. It would thus be highly interesting to exploit the possibility to grow superconducting materials on the present surface.

In contrast to S1/S2, the spin polarization of S3 could not be identified unambiguously. As shown in detail in figure 4, the apparent spin polarization is less than 0.1 at maximum for an extensive set of spectra for different angles and polarization conditions. We thus conclude that there is no obvious Rashba splitting for S3 within the experimental accuracy. The small but varying apparent polarization signal is thought to be due to the final state effect combined with the light polarization. We also carefully checked the high-resolution spin-intergrated ARPES spectra for the possible energy and momentum splitting of the S3 band (see figure 5). This also consistently supports that the Rashba splitting is smaller than the energy and momentum resolution of the experiment. These resolutions, for example, about 0.1 $Å^{-1}$, are mainly imposed by the spectral width of the S3 band itself. Note that the double layer model expected a Rashba splitting for S3, which is about half of that for S1/S2, that is, about 0.2 $Å^{-1}$ [22]. The present experiment indicates that the splitting, if any, should be much smaller than this theoretical expectation.

Although there is a consensus on the atomic origin of the half-filled bands, the origin of S3 is contrasted between different structure models. The calculation based on a single-row model suggests that S3 is due to the Si dangling bonds on step edges [13, 19], whereas that with the double-row model assigned it because of the second Au row [30, 32]. In this case, S1/S2 and S3 would be the bonding-antibonding pair for two Au rows. In an experimental point of view, the one-third-filled band was suggested to be related to Si step-edge chains, where the ×3 distortion is observed at low temperature [13, 16]. More recent STS studies denied the gap opening [26, 27]. Our own ARPES measurement, to be discussed elsewhere, also confirmed that S3 is metallic in the low temperature phase. This requests to discard the original claim of the Peierls mechanism for S3 and step-edge chains by Ahn *et al* [13, 16]. On the other hand, as discussed previously the Rashba splitting of S3 expected from its Au origin is not clear in the experiment. This leaves some question on the double-row model. Moreover, this model predicts a rather flat band for Si step-edge atoms at about 0.2 eV below Fermi level, which was not observed in ARPES and our own STS measurement [26]. These remaining discrepancies between the model

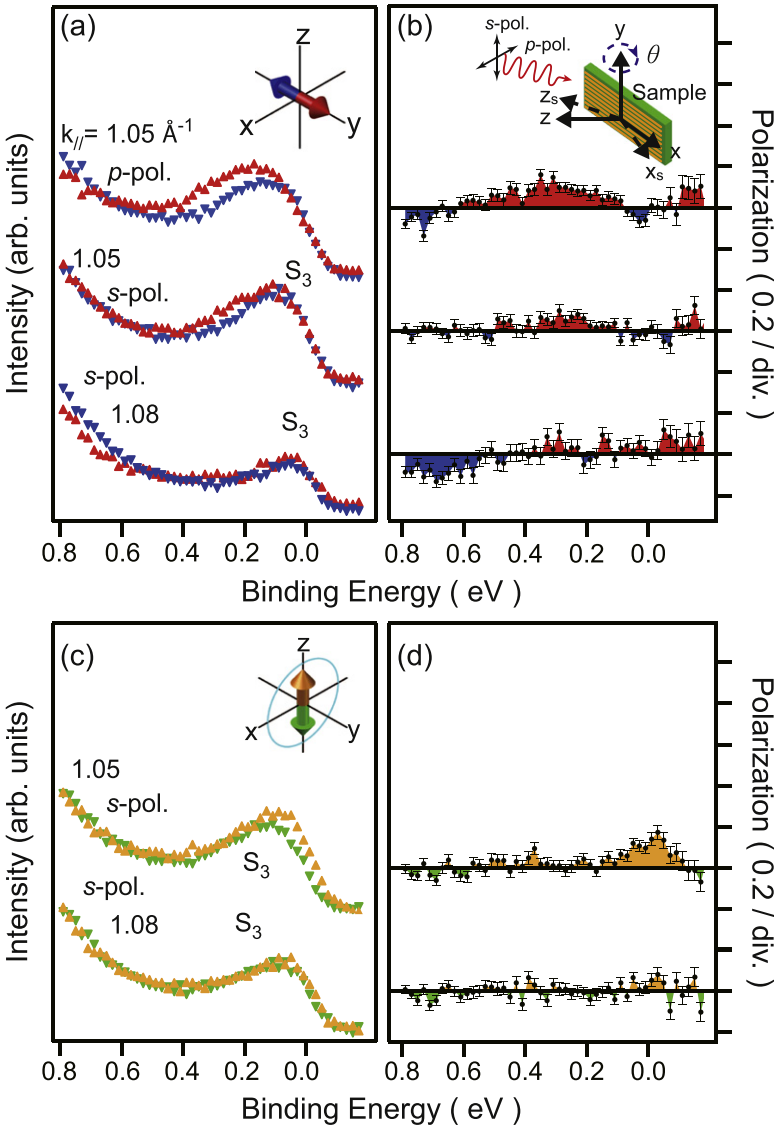

Figure 4. (a) and (b) Similar SARPES data as figure 2 but for the *k*-points showing the S3 state. The spin orientation is along k_y and the data are taken for two different light polarizations of *s*- and *p*-polarization, with respect to the surface plane. (c) and (d) Similar SARPES data but for the out-of -lane spin orientation. Note that the vertical scale of (b) and (d) is twice as large as those in figures 2 and 3.

and the experiments must be refined further before the structure model and the mechanism of the phase transition are fully understood.

Although the antiferromagnetic spin ordering and the Rashba splitting of Au-derived metallic bands provide new insight into the present system, they raise important questions, too. At present, the connection between the ×2 and ×3 transitions is not clear, as it was suggested they have different transition temperatures [16, 23]. More importantly, the nature of the ground state of the Au chains is not clear either. These metallic chains can be in the Luttinger liquid state, but the Rashba spin splitting makes the situation much more complicated. A very recent theoretical study showed that a two band system with a Rashba spin splitting can be in various

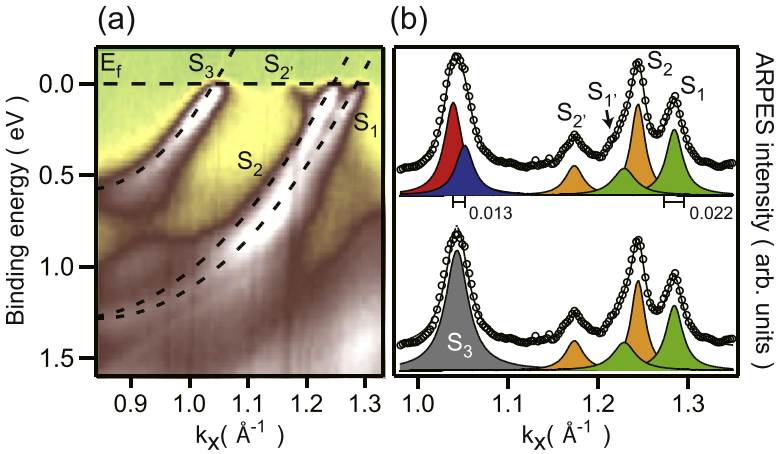

Figure 5. (a) High-resolution spin-intergrated ARPES data and (b) the momentum distribution curves of the ARPES data at the Fermi energy. These curves wave fitted with Lorentzians. The S3 state can be decomposed with double (top) or single (bottom) Lorentzians. The double Lorentzian fit is not unique with the peak positions, depending rather arbitrarily on the peak widths. That is, the splitting of S3, if present, must be smaller than the spectral resolution of the experiment.

different ground states with gapped spin or charge excitations and only part of the phase space is occupied by the Luttinger liquid state with gapless excitations [38]. We note that the real situation for the present system is even more complicated with the presence of yet another band S3. A very low temperature spectroscopy study for the present system would thus be very important together with further theoretical investigations.

4. Conclusions

We report on the direct experimental observation of the Rashba-type spin splitting using spin-resolved ARPES measurements for the 1-D metallic bands of atomic wires formed by Au adsorbates on a vicinal silicon substrate of Si(553). As proposed in the previous theoretical and experimental studies, the nearly half-filled doublet bands of S1 and S2 exhibit both in- and out-of-plane spin polarizations, but mainly in-plane polarization. This polarization can be explained by Rashba spin splitting. The other 1D metallic band S3 with a one-third filling does not show a substantial spin splitting within the experimental accuracy. Although these data are largely consistent with the recent Au double-row model and the antiferromagnetic spin ordering at low temperature, the refinement of the model for the electronic band structure is required. The large energy splitting of the spin-split bands, as large as about 0.25 eV, and the 1D nature of the conduction channel on a semiconductor substrate, in particular, on a silicon substrate, make the present system very attractive for spintronic applications and the search for a Majorana Fermion and the exotic manybody ground states.

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